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population to build up, (2) reduces population inversion between the first vibrational state and the ground state, and (3) introduces discharge instabilities that severely limit the pulse-rate capabilities. Average powers are reported to vary linearly with pulse-repetition rate up to 400 Hz, with typical pulse energies of 10 and 4 mJ for **small HF and DF** lasers respectively with discharge lengths of only 10 cm.

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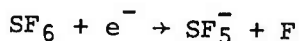
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1. INTRODUCTION

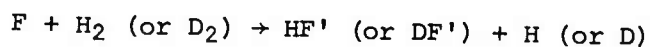
Electrically initiated chemical lasers offer promise for use in military systems because of their high energy-density output, high gain (allowing them to be made small), general simplicity of operation, and operation in wavelength regions of interest to the military.

Most work on pulsed chemical lasers has had the aim of producing large outputs at high efficiencies at low repetition rates.¹⁻³ Our aim has been to determine what output power and pulse energy can be obtained from small laser structures, to determine whether or not they can be operated at high repetition rates, and to measure their gross spectral characteristics at both low and high repetition rates.

The chemical reactions that we have used to produce the population inversion in vibrationally excited hydrogen fluoride (HF) and deuterium fluoride (DF) involve SF₆ as a fluorine-atom donor in the electron-attachment dissociative reaction⁴



followed by the reaction



¹H. Pummer and K. L. Kompa, Investigation of a 1-J Pulsed Discharge-Initiated HF Laser, *Appl. Phys. Lett.* **20** (1972), 356.

²R. A. Gerber and E. L. Patterson, Intense Electron-Beam Initiation of a High-Energy Hydrogen Fluoride (HF) Laser, *IEEE J. Quantum Electron.* **QE-10** (1974), 333.

³C. P. Robinson, R. J. Jensen, and A. Kolb, 60-J Pulses from an Electron-Beam-Initiated SF₆-C₂H₆ Chemical Laser, *IEEE J. Quantum Electron.* **QE-9** (1973), 963.

⁴D. C. Frost and C. A. McDowell, Electron Capture Processes in the Hydrogen Halides, *J. Chem. Phys.* **29** (1958), 503.

Hydrogenated carbon compounds such as ethane (C_2H_6) or propane (C_3H_8) can be used instead of hydrogen⁵ and were found to provide more stable operation without significant disadvantages other than forming deposits on electrode surfaces. Most of the work reported here on HF lasers used SF_6/C_2H_6 mixtures. Oscillation in DF was produced from SF_6/D_2 mixtures. Like DF, HCl has laser lines in the 3.5- to 4.2- μm atmospheric window,⁶ but handling problems and difficulties in obtaining acceptable power levels with Cl_2/H_2 (compared with an SF_6/D_2 mixture) argued against an extended effort to investigate this chemical laser system.

2. EXPERIMENTAL DETAIL

2.1 Laser Construction

A cross section of the resistor-loaded transverse-discharge laser structure used for spectral measurements is shown in figure 1. It consists of a 5-cm i.d. lucite tube equipped with two diametrically opposing rows of 51 (later 65) 1/2-W carbon resistors. The combined resistance of each pair of opposing resistors is 130 ohms. The spacing between the resistors is 0.41 cm and the anode-to-cathode spacing of the tips of the resistor pins is 1.1 cm. The two Mylar sheets, with perforations in line with each resistor pair, serve to equalize the gas flow. A series of 570-pF doorknob capacitors is attached alongside the upper (cathode) resistor array, one for every eight resistors. The tube is closed by detachable endpieces fitted with calcium-fluoride windows at the Brewster angle. The cavity consists of two high-reflectivity mirrors, one with a 312-cm radius and the other flat with a 1-mm output-coupling hole. Because of gas-flow limitations, this structure was usable only at very low pulse-repetition rates (up to a few hertz).

⁵T. V. Jacobson and G. H. Kimbell, *Transversely Pulsed-Initiated Chemical Lasers: Preliminary Performance of the HF System*, Chem. Phys. Lett. 8 (1971), 309.

⁶M. A. Pollack, *Molecular Gas Lasers*, in *Handbook of Lasers*, R. J. Pressley, ed., Chemical Rubber Company, Cleveland, Ohio (1971), 298-349.

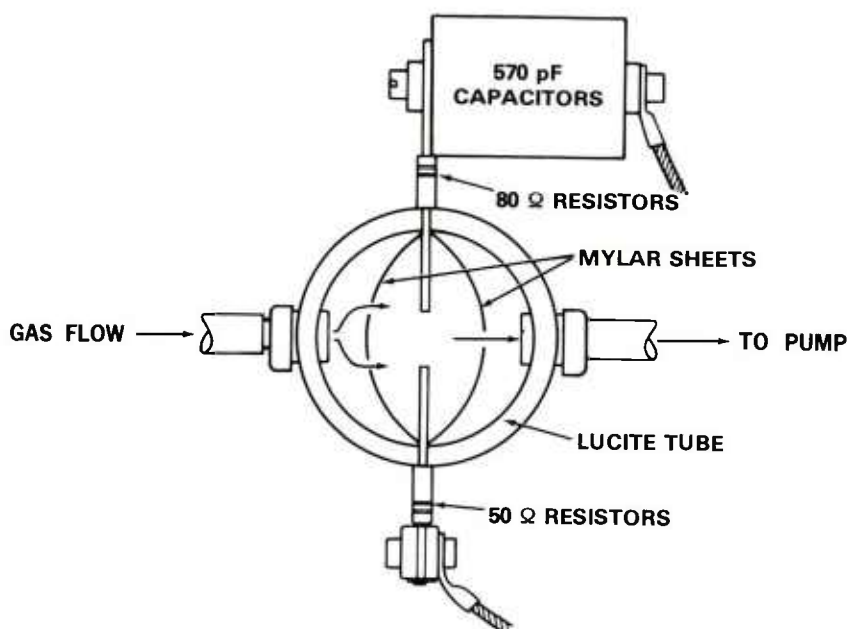


Figure 1. Cross section of resistor-loaded laser structures used for low repetition-rate measurements.

For producing more output and for operation at higher repetition rates, another construction was chosen. It was designed for a higher gas-flow rate and allowed for easy exchange of or modification to the electrode assemblies and gas manifolds. The various parts were held together with screws (instead of cement) and the seals were made with O-rings. Figure 2 shows a cross section of this structure. The cathode-pin array is 10 cm long and contains 30 pins each in series with a carbon 100-ohm 1-W resistor. The pins were bent to provide a linear array of points in line with the anode. The cathode-series resistors were cooled with a stream of compressed air. The anode consisted of a sharp metal edge flush with the inner Teflon surface. Dead space, where combustion products could be trapped, was kept to a minimum and efforts were made to achieve an uninterrupted gas flow. Other electrodes such as non-resistor-loaded Rogowski-profile configurations, with and without pre-ionization, were also tried. These were found to give a smaller output and had a greater tendency to arc formation at all repetition rates. The output mirror used with this laser structure was dielectric coated and had a reflectivity of 30 percent at 2.8 μm .

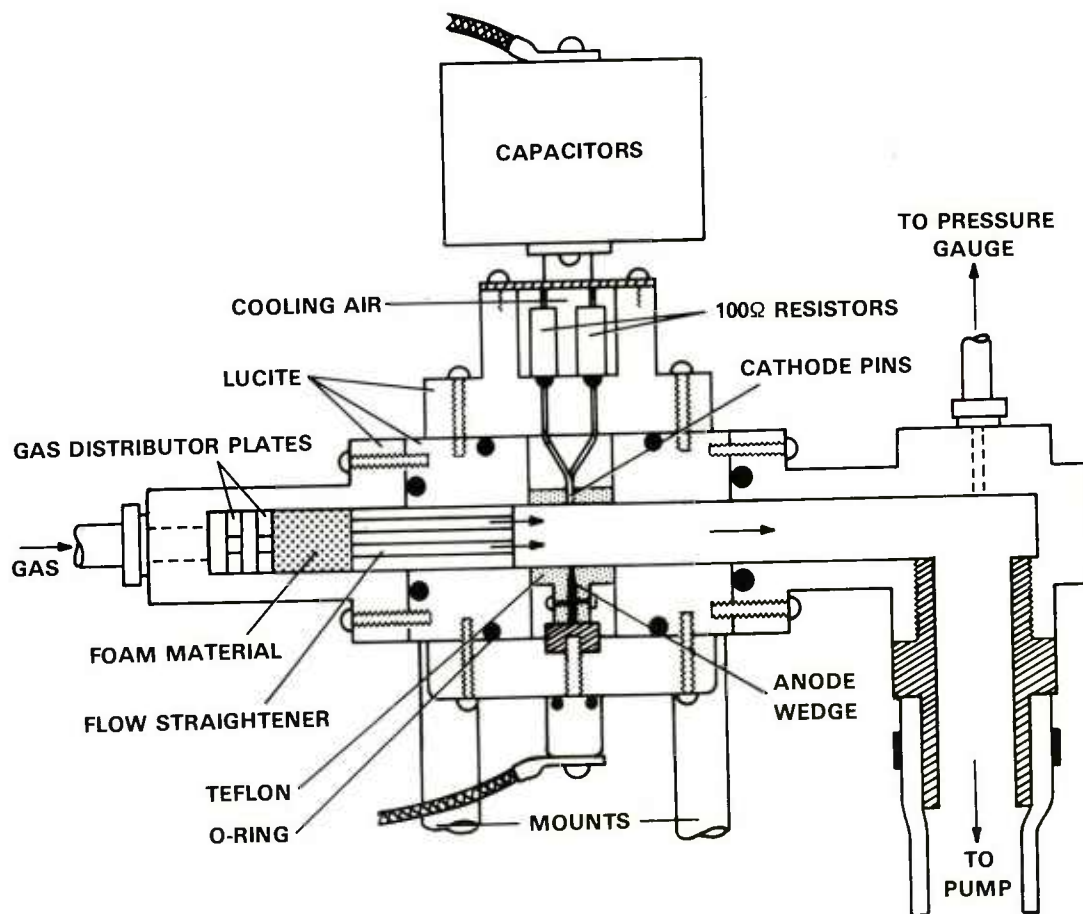


Figure 2. Cross section of demountable laser structure allowing for faster gas flow and usable at high pulse-repetition rates.

Also constructed were a number of very small longitudinally and transversely excited laser structures with a discharge length of only 2 cm. The mirrors were mounted directly onto the laser structures. Output coupling was through a small hole in one mirror that was sealed with a small piece of CaF_2 or thin plastic foil. These miniature lasers functioned well with an $\text{SF}_6/\text{C}_2\text{H}_6$ mixture to give outputs no greater than 0.5 mJ. Only the strongest lines that were produced with the larger lasers occurred with these miniature lasers.

2.2 Laser Electrical Circuitry

2.2.1 Low Repetition Rates

Figure 3 shows the discharge circuitry that was used at repetition rates of a few hertz. The circuitry is simple, and because one side of the spark gap and the anode are at ground potential, high-voltage isolation problems and rf interference from corona are minimized. The laser structure is shunted by a conductive element, in this case a resistor, so that capacitor C can be charged.

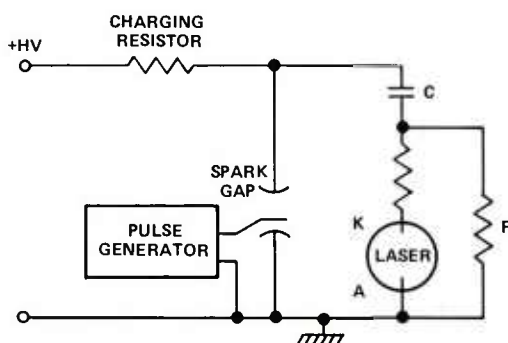


Figure 3. Electrical circuit used with low repetition-rate lasers.

2.2.2 High Repetition Rates

To achieve high repetition rates, the spark gap was replaced by an EG&G HY-32 hydrogen-filled thyatron (fig. 4). Quenching problems with the thyatron/laser combination could not always be overcome simply by adding a negative bias to the grid. We concluded that to turn off the thyatron the current had to have its direction reversed or at least be made to go to zero. This current reversal usually can be achieved by using a small inductance in the anode circuit of the thyatron. Such an insertion, however, increased the rise time of the current pulse and adversely affected the laser output. A small inductor L across the laser channel in place of the charging resistor R in figure 3 was found to improve quenching of the thyatron. Its performance is illustrated in the following way. If the charge voltage is too small to produce a discharge in the laser channel when the thyatron fires, the inductor L takes the whole current. In this situation, shown in figure 5a, after a half-period ($\pi\sqrt{LC}$) when the capacitor C has reversed its polarity and the current goes through zero, the thyatron opens and the circuit oscillates with the shorter half-period $\pi\sqrt{LC'}$ where $C' = CC_1/(C + C_1)$, with C_1 the capacitance of the thyatron (fig. 5, b and c). When the voltage is set sufficiently high to produce a discharge in the laser

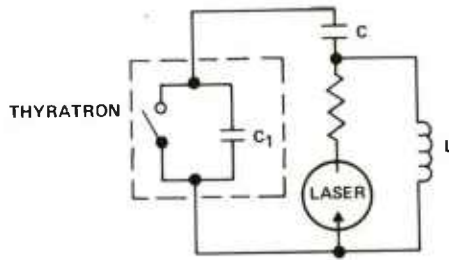


Figure 4. Equivalent circuit incorporating thyatron and inductance shunt for high repetition rates.

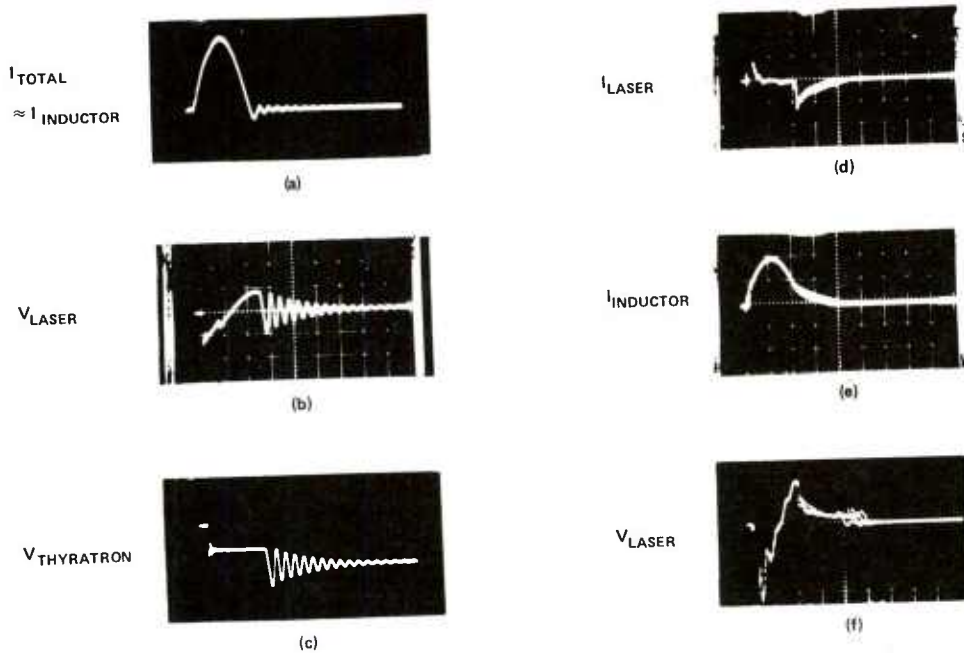


Figure 5. V-I characteristics of laser circuitry (shown in fig. 4): (a), (b), (c), at 5-kV charge voltage, laser not firing; (d), (e), (f), 10-kV charge voltage, laser firing. Inductance value: $88 \mu H$; time scale: 1 division equals $1 \mu s$.

channel and the initial current pulse during which lasing occurs does not completely discharge capacitor C, then the circuit consisting of the laser channel and L begins to oscillate. Shortly before the half-period is over, the reversed voltage across the laser is large enough to produce another discharge (usually an arc discharge) in the laser channel (fig. 5d). This second discharge pulse, which is a reverse current through the laser, adds a tail to the otherwise sinusoidal current through the coil (fig. 5e). Voltage irregularities that are observed shortly before the end of this process may be an indication of small space-charge buildups that tend to suppress the current (fig. 5f). Even though most of the charge on the discharge capacitor C flows through the laser cavity, the small reverse current is able to quench the thyatron. With higher operating voltages, the current distributions become more complex, and nonquenching of the thyatron occurs more often, and the laser output continues to increase with increasing charge voltage.

2.3 Maximizing Laser Output

The total pressure that gave the largest output at charge voltages of about 20 kV was found to be approximately 40 Torr (5.3 kPa) in all cases. This pressure increased slightly with higher charge voltages. Sulfur hexafluoride is always the majority component in the gas mixture. Only small amounts of hydrogenated carbon compounds, hydrogen, or deuterium are required (in the approximate ratio of 1:20 with SF₆). Deuterated compounds were not used because of their expense. Maximization of the laser output required careful adjustment of the minority gas pressure. This requirement appeared to be due more to the effect of the minority gas pressure on the discharge and its tendency to introduce arc discharges that terminate laser action than to the introduction of deactivation processes that would reduce the laser output.

Uniform and laminar gas flow in the transverse-discharge region was found to be very important. The output pulse energy was more than doubled after a 15-mm layer of porous plastic foam material was installed in the inlet manifold followed by flow straighteners ending about 12 mm from the electrodes. The gas flow was made to extend beyond the ends of the discharge region. To reduce turbulence, and to try to avoid the accumulation of discharge products in the discharge region, the wedge-shaped anode was made flush with the surface. When a pin array was used instead of the wedge-shaped anode, it was made to protrude only 1 mm from the Teflon surface. This protrusion was found to be a practical necessity to avoid erosion of the Teflon surface by the individual discharges.

At high repetition rates, the overall discharge tended to concentrate at those electrodes that were closest to the points where the capacitors were connected. It was found to be advantageous, therefore, to make up the total discharge capacitance with several small capacitors and distribute them along the resistor-loaded pins to minimize discharge nonuniformity. This procedure improved the pulse energy by 30 percent.

Arcing has a deleterious effect on the laser output, especially in operation at high repetition rates. Arcing is a secondary nonlasing localized discharge driven by a residual voltage on the capacitor that has been left over from the main diffuse (laser) discharge. This behavior appears to be characteristic of discharges containing electronegative gases like SF_6 where heavy negative ions (SF_6^- or SF_5^-) build up a space charge and suppress the current flow.⁷ After a period of time, which can last from a fraction of a microsecond to several tens of microseconds, the space charge has dissipated so that current flow is again possible. Unfortunately, the current flows in a single or double nonlasing arc. The effect of the arcing can be reduced in severity (but not in frequency of occurrence) by shunting the laser channel with a resistor or inductor that has a value which is small enough to provide an RC time constant that enables any voltage remaining on the capacitor to be quickly reduced between excitation pulses. Surprisingly, unlike others working with pulsed CO_2 lasers,⁸ we found that the residual voltage on the capacitor could not be reduced enough to avoid initiating an arc discharge. Figure 6 shows a typical V-I (voltage-current) characteristic of a small HF laser, illustrating this behavior clearly. The voltage characteristic rises sharply to a high voltage following firing of the spark gap (or thyatron), then falls even more sharply, and tapers off to a plateau that is typically one half to one third of the peak voltage. While the voltage is falling, the diffuse-discharge current pulse occurs. It has a typical full width at half maximum (FWHM) of about 70 ns. During the same period, the laser pulse begins. The laser pulse continues beyond the time at which the current pulse has terminated and has a typical FWHM of approximately 100 to 150 ns.

⁷J. D. Cobine, *Gaseous Conductors*, Dover Publications, New York (1958), 264.

⁸Y. L. Pan, A. F. Bernhasdt, and J. R. Simpson, *Construction and Operation of a Double-Discharge TEA CO_2 Laser*, Rev. Sci. Instrum. 43 (1972), 662.

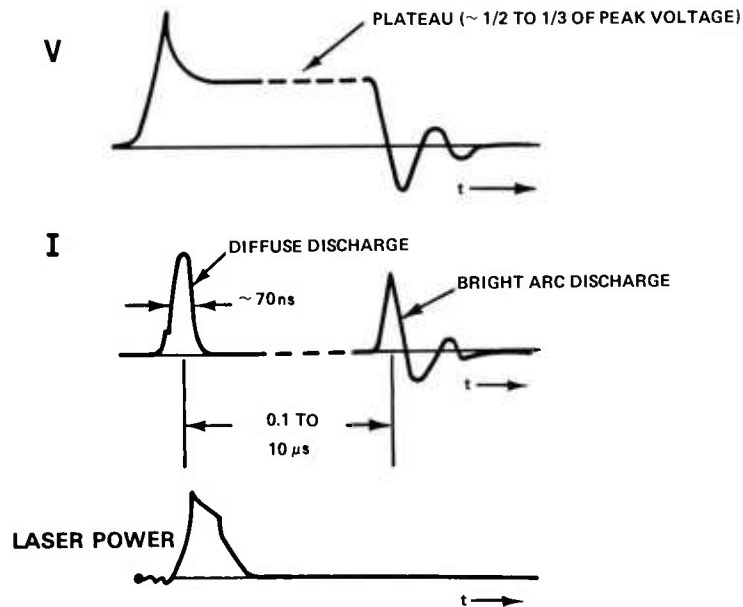


Figure 6. Correlation between voltage, current, and laser output. Non-resistor-loaded HF laser structure; C_2H_6/SF_6 mixture.

The V-I characteristic of figure 6 also shows the relationship between the diffuse-discharge pulse and the typical (ringing) current pulse of a bright arc discharge. When the tendency for arcing is high, the arc-discharge current pulse moves closer to the diffuse-discharge pulse and is sometimes hardly distinguishable from it. It is only when the localized arc pulse occurs almost simultaneously with the diffuse-discharge pulse that it terminates the laser output pulse. This termination appears to occur mainly through localized distortion of the optical cavity. Seldom does the arc discharge occur soon enough following the breakdown of the laser mixture to prevent the formation of the diffuse discharge. If the charge voltage is increased, it is found that the residual-charge voltage decreases, and the magnitude of the bright arc-discharge current pulse is also decreased. Presumably, the increased laser output at higher charge voltages reflects better utilization of the available energy stored in the charge capacitor rather than the effect of a higher E/p (E is electric field and p is pressure). This behavior--the occurrence of the laser pulse only during the period when the laser voltage is falling rapidly--is unlike that reported for pulsed CO_2 lasers, with which the laser pulse occurs for

a longer period in which the voltage is falling so slowly that one can meaningfully talk about a constant E/p period.^{9,10} The rapid fall in voltage during the diffuse-discharge current pulse is presumably caused by strong electron attachment in the presence of SF₆ and its discharge products.

2.4 Measuring Techniques

The average output power was measured with a disc calorimeter in combination with an electronic power meter. This meter was later replaced by a needle microammeter to avoid rf interference at high power measurements when the laser is operating at high repetition rates. The laser pulse energy was calculated from measurement of the average output power and the known pulse-repetition rate. Various pulsed voltages and currents were monitored with a high-voltage probe and a current transformer, respectively, each with rise times of approximately 50 ns. For spectral measurements at the exit of the 1/2-m spectrometer, a liquid-nitrogen-cooled indium-antimonide photovoltaic detector was used connected to an amplifier. For recording purposes, the pulsed signals were rectified, further amplified, and matched to a strip-chart recorder. Since the amplifier did not respond in the same way to pulses of differing rise times and shapes, the relative magnitudes of individual pulses are not meaningful, and the detector-amplifier setup merely provided a means of detection of the presence of radiation. Actual pulse shapes of individual lines were obtained with a liquid-helium-cooled copper-doped germanium photoresistor and were displayed directly on a fast oscilloscope. The rise time of this combination was less than 5 ns.

3. RESULTS

3.1 Spectra at Low Repetition Rates

Spectra were obtained for HF and DF lasers of the low gas-flow, low repetition-rate type shown earlier in figure 1. The pressure and gas composition were optimized for producing the highest total pulse energy. The HF laser transitions usually obtained with SF₆ and ethane were P₁(7 to 12), P₂(3 to 12), and P₃(3 to 9) except for P₁(10) and P₂(10). Other lines, such as P₂(16), also occur for certain narrow

⁹L. J. Denes and J. J. Lowke, V-I Characteristics of Pulsed CO₂ Laser Discharges, *Appl. Phys. Lett.* **23** (1973), 130.

¹⁰O. P. Judd and J. Y. Wada, Investigations of a UV Preionized Electrical Discharge and CO₂ Laser, *IEEE J. Quantum Electron.* **QE-10** (1974), 12.

excitation conditions and cavity alignment, both of which required careful optimization. With smaller lasers, the spectrum shifts to transitions with lower rotational numbers P_1 (4 to 8), P_2 (3 to 9), and P_3 (3 to 8). With SF_6 and hydrogen mixtures, we observe P_1 (12 to 16), P_2 (3 to 9, 11 to 13, and 16), and P_3 (5 to 6) HF laser lines. Only the uninterrupted series P_2 (7 to 12) was observed by using a mixture of freon 12 and hydrogen. It can be concluded that the use of SF_6 is responsible for the nonoccurrence of the P_2 (10) HF laser line. In contrast to the limited spectra obtained with mixtures of hydrogen and SF_6 , a mixture of deuterium and SF_6 produced as many as 36 DF laser lines in the four series P_1 (8 to 16), P_2 (4 to 14), P_3 (4 to 14), and P_4 (6 to 11).

3.2 Spectra at High Repetition Rates

High repetition-rate spectra were obtained with the laser structure (shown in fig. 2) that was designed specially for fast gas flows. The HF and DF laser lines were observed by using mixtures of SF_6 with ethane, hydrogen, or deuterium. The spectrum is the same as that for operation at low repetition rates as long as there is enough time between successive pulses to restore the original discharge conditions. This is the operating region where the average output power increases linearly with increase in pulse-repetition rate. For times shorter than the gas-transit time, the spectrum shrinks, starting at the P_1 and P_3 series. This occurs at pulse-repetition rates of between 300 and 500 Hz for $\text{SF}_6/\text{C}_2\text{H}_6$, 200 Hz for SF_6/H_2 , and between 200 and 400 Hz for SF_6/D_2 laser mixtures (as shown in fig. 7).

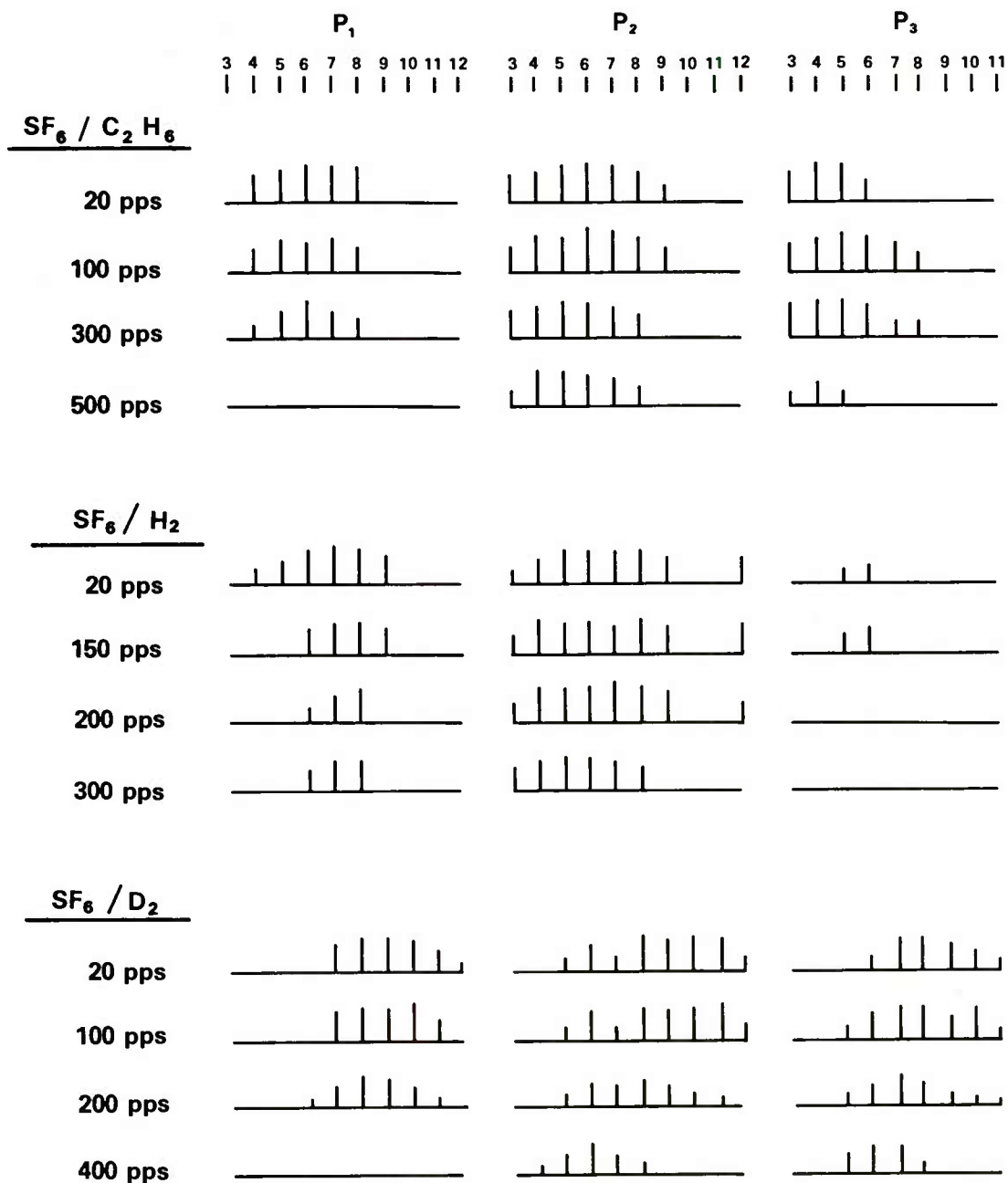


Figure 7. HF and DF laser spectra at increasing pulse-repetition rates.

3.3 Laser Pulse Duration

The duration of the individual HF laser lines varied from 30 ns for the shortest-duration lines to 170 ns for the longest--P₂(8). The delay of oscillation on transitions with higher rotational numbers becomes more pronounced when the pulse-repetition rate is increased (fig. 8). With SF₆ and H₂ instead of C₂H₆, the transitions appear to belong to two groups: one with rotational numbers below, and another with rotational numbers above 10. Only the latter group, J = 11 to 13, seems to cascade down from vibrational state 2 to the ground state.

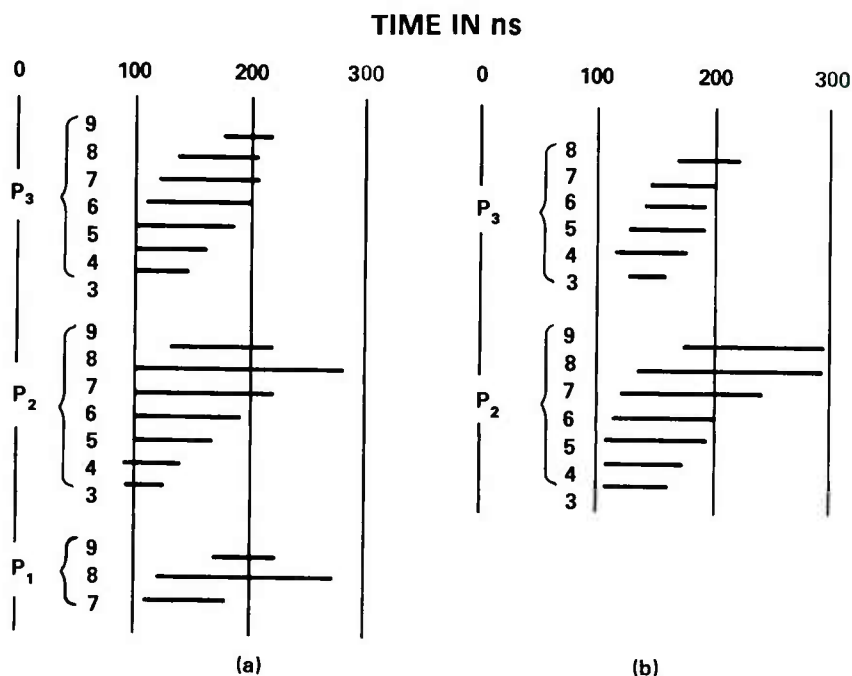


Figure 8. Pulse duration and relative time of occurrence of individual HF laser lines from a C_2H_6/SF_6 mixture observed at (a) 1 pulse/s and (b) 12 pulses/s.

3.4 Output Power/Pulse Energy

3.4.1 At High Repetition Rates

The power output up to repetition rates of approximately 1 kHz was measured for optimized mixtures of SF₆ with ethane, hydrogen, and deuterium with charging voltages of 20 kV with the various capacitance values indicated. The results are shown plotted in figures 9 to 11. The optimum total pressure was about 40 Torr (5.3 kPa), and the best mixture ratio was about 20:1 with SF₆ the main component. The exact ratio was always adjusted at the beginning of each run because the output was sensitive to the mixture ratio. The gas-flow rate at this pressure was between 300 and 600 cm/s. The average power is first linear with pulse-repetition rate up to approximately 200 Hz, becomes less than linear, and finally drops off sharply between 400 and 1000 Hz depending on the capacitance value. Beyond the maximum, the discharge is heavily arcing and reproducible measurements are not possible.

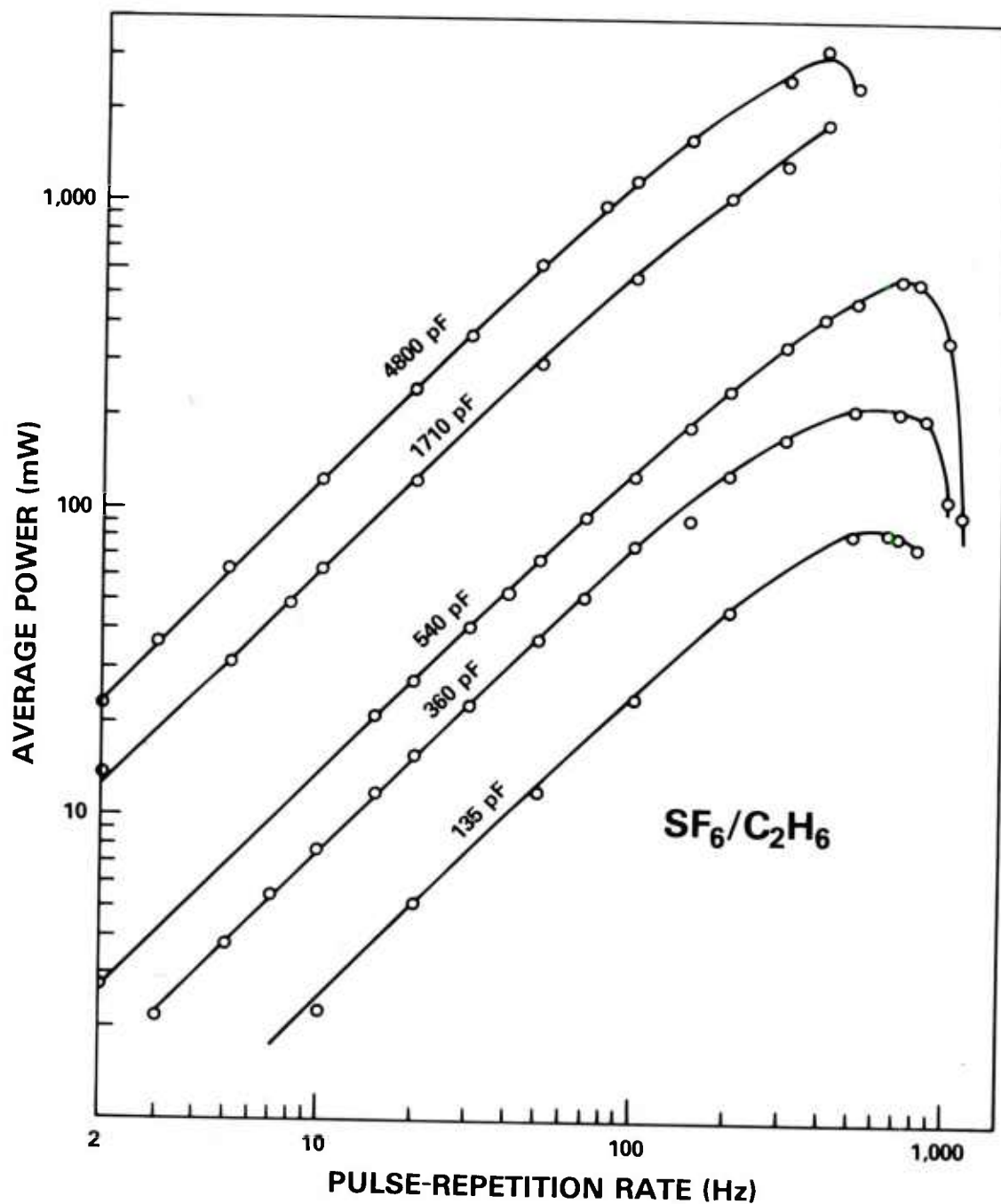


Figure 9. Average HF-laser output power versus pulse-repetition rate with an $\text{SF}_6/\text{C}_2\text{H}_6$ mixture for various values of capacitance; fast gas flow.

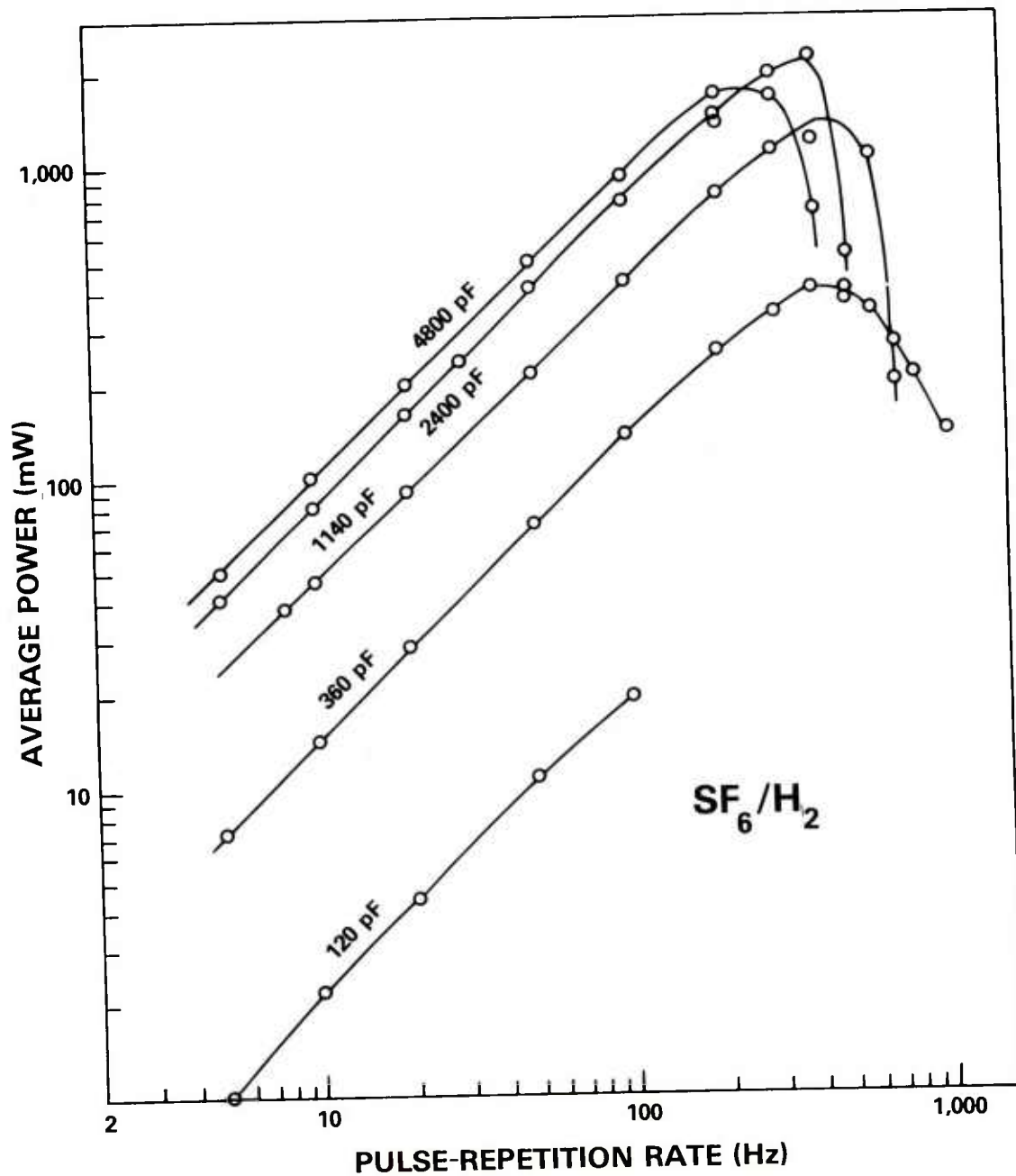


Figure 10. Average HF-laser output power versus pulse-repetition rate with an SF_6/H_2 mixture for various values of capacitance; fast gas flow.

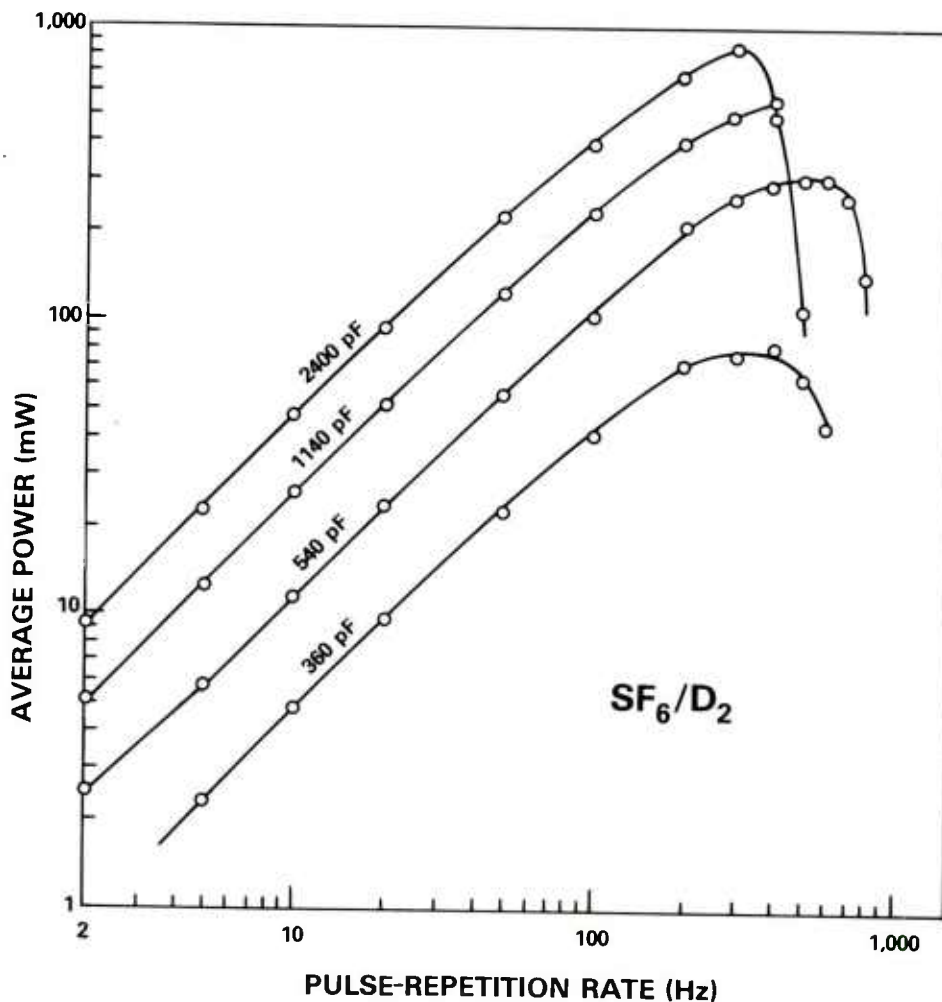


Figure 11. Average DF-laser output power versus pulse-repetition rate with an SF₆/D₂ mixture for various values of capacitance; fast gas flow.

3.4.2 Dependence on Capacitance

The energy per pulse was measured for various values of discharge capacitance with charge voltage kept constant at 20 kV. The measurements were taken only in the low repetition-rate region where the output is directly proportional to the pulse frequency. Despite attempts to optimize the conditions for each measurement, considerable inconsistency and spreading of the data points were experienced. As illustrated in the double-logarithmic plots of figures 12 and 13, the pulse energy increases more than linearly with the capacitance (approximately proportional to $C^{1.4}$). At capacitance values above 2400 pF, the pulse energy deteriorates. This is especially the case

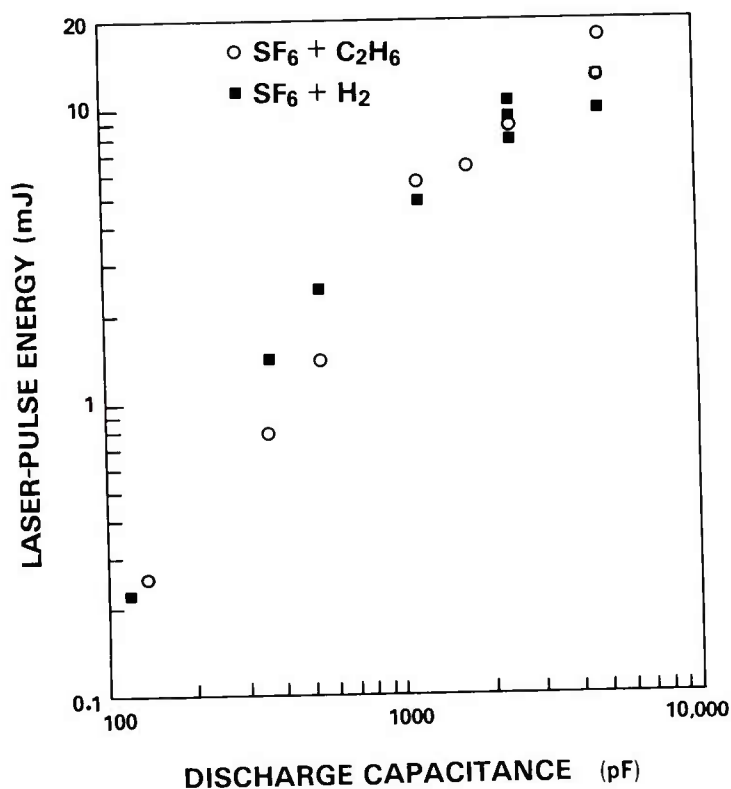


Figure 12. Laser-pulse energy as a function of capacitance (20-kV charge voltage) when operated as an HF laser.

with SF_6 and D_2 above 2400 pF, when arcing dominates over diffuse-discharge formation. The pulse energy for a particular capacitance value is almost a factor of 2 larger for HF than DF. There is a spread to the value of the pulse energy versus discharge capacitance with both $\text{SF}_6/\text{C}_2\text{H}_6$ and SF_6/H_2 mixtures, and no clear difference is apparent between the output energies realizable from either laser mixture. The maximum pulse energy realized for HF laser mixtures was approximately 15 mJ for a discharge capacitor of 4.8 nF, and 6 mJ per pulse from a DF laser mixture for a 500-pF charge capacitor.

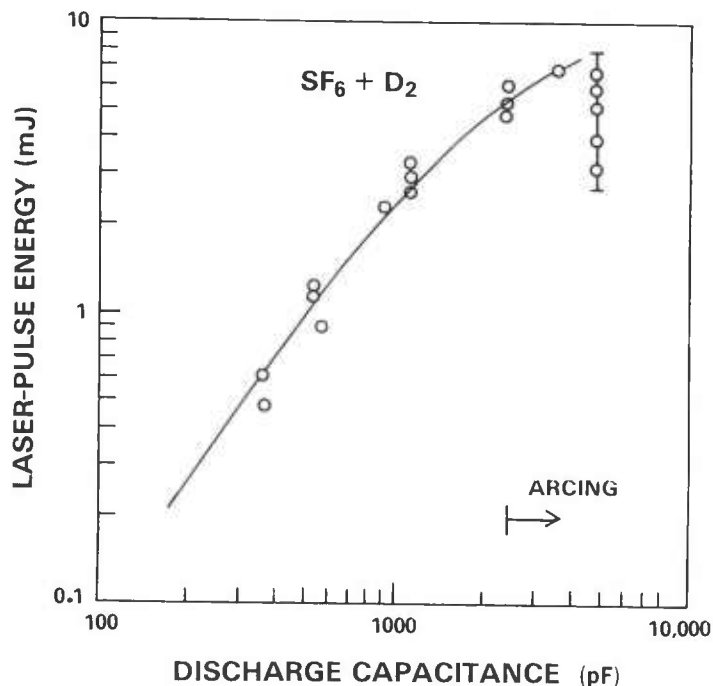


Figure 13. Laser-pulse energy as a function of capacitance (20-kV charge voltage) when operated as a DF laser.

4. DISCUSSION AND CONCLUSIONS

We have determined the gross spectral characteristics and the output behavior of small HF and DF lasers operating at pulse-repetition rates up to 1 kHz. At this repetition rate, the maximum pulse energy that we have been able to achieve with HF was approximately 0.4 mJ. With DF, we achieved a repetition frequency of 800 Hz with a 0.1-mJ pulse energy. At these high repetition rates, heavy arcing occurs with wide fluctuations in the laser output. Heavy arcing, in all the lasers that we investigated, caused the deviation from linearity between average output power and pulse-repetition rate that occurred at lower repetition rates, as illustrated in figures 9, 10, and 11.

All our measurements and observations indicate that the diminishing laser output at high repetition rates is due to an insufficient gas-flow rate and possibly nonuniformity in the gas flow. The insufficient gas-flow rate is responsible for two effects that reduce the energy per pulse at high repetition rates. The two effects are (1) buildup of

population of HF and DF molecules in their zero-vibrational ground states, and (2) nondispersion of long-lived excited or ionized reaction products in or close to the discharge region or slightly downstream of the discharge region.

The first effect, buildup of population in the ground state, is responsible for the preferential disappearance of oscillation in the P_1 transitions to the ground state at high repetition rates (as evidenced by fig. 7).

The second effect (evidenced by heavy arcing and the behavior of the V-I pulsed characteristics of the lasers) alters the excitation conditions from pulse to pulse at the high repetition rates and produces nonuniform excitation in the discharge region. At high repetition rates, the normal spraylike discharge between electrodes assumes more the appearance of a flame that is blown downstream to one side of the discharge region. During the short time of one pulse, the gas moves only a distance of 30 μm so that it can be considered stationary in that time. The flamelike glowing, or arcing on the downstream side, therefore, must be due to long-lived ions or excited species generated from previous pulses that cause the discharge to take a path that is out of the laser-cavity region, and which has the wrong conditions for efficient dissociation of SF_6 to the required free F-atoms (and SF_5 ions).

The apparent change in excitation conditions at high repetition rates was not due to power limitations in the high-voltage power supply that would cause the discharge capacitors to be incompletely charged between pulses. At the higher repetition rates, the generator voltage was raised to maintain constant the charge voltage on the capacitors and the amplitude of the discharge current pulses.

Our gas-flow velocity in the discharge region was approximately 500 cm/s, calculated from measurements of the gas flow and pressure at the exhaust of the vacuum pump and from measurement of the pressure in the discharge region. On the assumption that the lateral extent of a single pin discharge is about 3 mm, this flow rate should ensure that gas is completely changed at pulse-repetition rates up to 1.6 kHz. Since arcing occurred in this work at about one third of this value, one would think that insufficient gas flow could not be invoked to account for nonlinearity in the output power at high repetition rates. It has been suggested, however,* that acoustical shock waves triggered by the

*G. S. Dzakowic and S. A. Wutzke, *The Influence of Transverse Flow upon High Pressure, High-Repetition-Rate Flow Discharges*, presented at the 25th Gaseous Electronics Conference, London, Ontario, Canada, (1972).

discharge can cause reaction products to be moved upstream against the flow direction. The effect of this would be to require that the gas flow be increased to offset the upstream movement to ensure that the gas in the discharge region is completely changed between excitation pulses.

Introduction of flow straighteners and of an impedance in the gas inlet to the laser (to improve the uniformity of the gas flow along the extent of the electrodes) did significantly improve the laser output power and slightly raise the arcing threshold to higher repetition rates. This improvement would imply that the uniformity of gas flow in the discharge region is of considerable importance in operating at high repetition rates.

Our results show that electrically initiated HF and DF lasers with active regions of approximately 10 cm can be constructed to give multiline output-pulse energies of approximately 10 and 4 mJ, respectively, at repetition rates of up to a few hundred hertz.

Flow rate and uniformity have been found to be of prime importance in achieving operation at high repetition rates. Insufficient flow rate and nonuniformity in the gas flow are responsible for a reduction in the laser output through buildup of population of laser molecules in the ground state, and through the introduction of arcing that alters the excitation conditions at high repetition rates. The tendency for arcing can be reduced but not eliminated by the use of resistor-loaded pin electrodes in a single linear array at right angles to the gas-flow direction.

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